The effect of rapid thermal annealing on the photoluminescence of InAsN/InGaAs dot-in-a-well structures

R Gargallo-Caballero , J Miguel-Sánchez , Á Guzmán , A Hierro and E Muñoz

Instituto de Sistemas Optoelectrónicos y Microtecnología (ISOM)–Departamento de Ingeniería Electrónica, ETSI Telecomunicación, Universidad Politécnica de Madrid, Ciudad Universitaria s/n, 28040 Madrid, Spain

Abstract
The effect of post-growth rapid thermal annealing on the optical characteristics of InAsN/InGaAs dot-in-a-well DWELL structures grown by molecular beam epitaxy on GaAs(1 0 0) has been studied. InAs/InGaAs DWELL structures have been used as a reference. Photoluminescence measurements of these samples show similar optical effects, such as a blueshift of the peak wavelength and a reduction of the full width of at half maximum PL emission, in both types of structures up to an annealing temperature of 750 °C. Nevertheless, at 850 °C, these effects are much more pronounced in the structures with N. These results suggest that an additional As–N interdiffusion process inside the InAsN quantum dots plays a dominant role in these effects at high annealing temperatures (850 °C) on InAsN/InGaAs structures.

1. Introduction

In the past few years, InAs/InGaAs quantum dot-in-a-well (DWELL) structures have emerged as a very important material system to tune the emission wavelength of optical communication systems at around 1.3 μm Using an InGaAs capping layer instead of the conventional GaAs layer leads to a reduction of the confinement potential for the carriers in the quantum dots (QDs) as well as a reduction of the strain in these nanostructures; therefore, the emission wavelength achieved is shifted to higher values. Another important technique to increase the emission wavelength is the growth of large InAs QDs which implies low growth rates ($\sim$0.01 ML s^{-1}) The use of an InGaAs capping layer together with the growth of such large InAs QDs simultaneously could extend further the emission wavelength but, in the case that the MBE system has only one indium cell which is quite usual, it involves employing some additional growth methods (e.g. growth interruptions [3]). In this case, because of the low growth rate for InAs QDs, the redshift achieved with large QDs is lost when the InGaAs capping layer is grown at such low growth rates, due to In segregation. One way to extend the emission wavelength using DWELL structures and avoiding these drawbacks is the incorporation of N into the QDs, i.e. the growth of InAsN/InGaAs DWELL structures, due to the strong reduction in the bandgap of the III–V semiconductors by the incorporation of small concentrations of atomic nitrogen. Moreover, an additional phenomenon contributes to the PL redshift. The incorporation of N into the QDs allows us to decrease the lattice parameter of the QDs, leading to a reduction of the compressive strain in the QDs. Similarly, we could avoid low growth rates to obtain large QDs and so In segregation, since N allows us to extend QDs' emission wavelength without the need for increasing their dimensions. Moreover, the growth of InAsN QDs can help in overcoming some disadvantages related to the increase in the dimensions of these nanostructures such as the formation of dislocations inside the QDs due to the huge strain accumulated in them. Thus, the incorporation of N into the InAs QDs of DWELL structures seems to be an advantageous option to redshift the emission wavelength to values as high as 1.55 μm, due to the strong reduction in the bandgap and in the compressive strain in the QDs.

Nevertheless, we have to take into account that for fabricating light-emitting devices based on these DWELL structures, we must increase the growth temperature after the growth of InAsN QDs and the InGaAs capping layer (i.e. to grow the GaAs region and the p-type contact). Also, the N normally degrades the optical properties of the structures in which it is incorporated, reducing the optical intensity of PL and EL. According to previous works of dilute nitrides it is possible to improve these properties performing conventional annealing or rapid thermal annealing (RTA) cycles of these samples. Furthermore, it will be very important to study the effect of post-growth RTA on InAsN QDs. The main objective of this work has been to investigate this effect on the optical characteristics of the InAsN/InGaAs DWELL structures using post-growth RTA treatment.

2. Experimental procedure

In this work, the samples were grown on GaAs(1 0 0) substrates using a MBE Riber 32 equipped with a radiofrequency (RF) Oxford Applied N Plasma Source. We have grown two different DWELL samples: one of them consisting of InAs QDs buried with InGaAs, and the other one consisting of InAsN QDs buried with InGaAs. Both these samples were grown under the same growth conditions to make a later comparison between them. QDs were grown with a nominal thickness of 4 ML at 0.15 ML s^{-1} and at 470 °C. Then, a capping layer of $In_{0.2}Ga_{0.8}As$ of 12 nm was used to cover the QDs at the same substrate temperature. For the QDs in the InAsN/InGaAs DWELL sample, we used a N plasma generated using 0.2 sccm of N_2 flux and a RF power of 65 W. The active nitrogen generated is monitorized by an optical emission detector (OED). In both cases, the growth of the buried QDs was followed by the growth of a 250 nm GaAs layer at 590 °C. The growth rate of GaAs was 0.6 ML s^{-1}. The formation and evolution of the growth were monitored by reflection high-energy electron diffraction (RHEED).

RTA cycles were carried out on both QDs samples in a N_2 ambient at 1.3 bar at temperatures ranging from 600 to 850 °C for 30 s. The samples were sandwiched between S-I GaAs wafers during annealing to reduce As desorption.

The optical characterization of these samples was carried out by photoluminescence (PL) measurements performed at low temperature (15 K) using a closed-cycle He cryostat under the excitation of a He–Ne laser. The PL spectra were acquired using a cooled Ge detector and a conventional lock-in technique.

3. Experimental results and discussion

Figure 1 shows the PL spectra from the two types of DWELL as-grown samples. As expected, with N incorporation into the InAs QDs, there is a strong reduction of the QDs bandgap with the result that there is a strong redshift in the wavelength of 193 nm. The peak wavelength at 15K obtained from the InAsN/InGaAs DWELL is 1.27 μm, very close to the technologically important 1.3 μm. Also, a broadening of the InAsN QDs PL full width at half maximum (FWHM) in comparison with the InAs QDs one is seen. Nevertheless, the integrated PL (IPL) intensity is similar for both cases. This

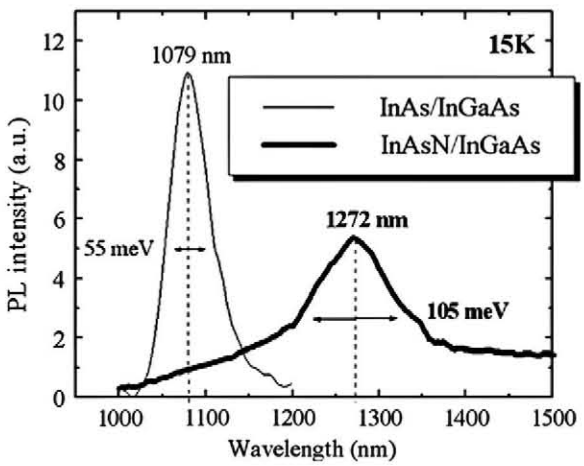

Figure 1. PL spectra at 15 K of as-grown InAs/InGaAs DWELL and InAsN/InGaAs DWELL samples grown under the same growth conditions.

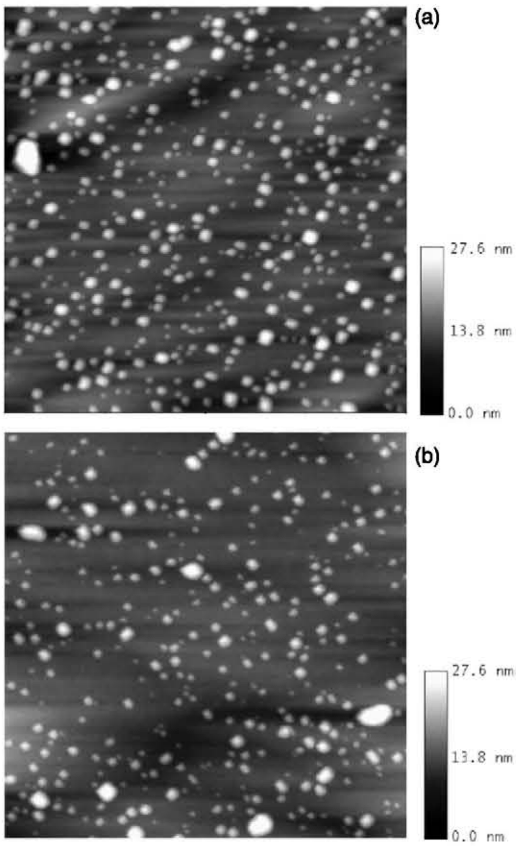

Figure 2. The $1 \times 1\ \mu m^2$ AFM images of (*a*) InAs and (*b*) InAsN QDs grown under the same growth conditions.
(This figure is in colour only in the electronic version)

effect could be induced either by a greater size dispersion of the QDs when they are grown with N and/or by an inhomogeneity of N concentration between the InAsN QDs, the usual characteristic in as-grown dilute nitride quantum wells grown by MBE In figures 2(*a*) and (*b*) $1 \times 1\ \mu m^2$ AFM images of InAs and InAsN QDs grown under the same growth conditions are shown, respectively. As we can observe in these images, the dimensions and density of

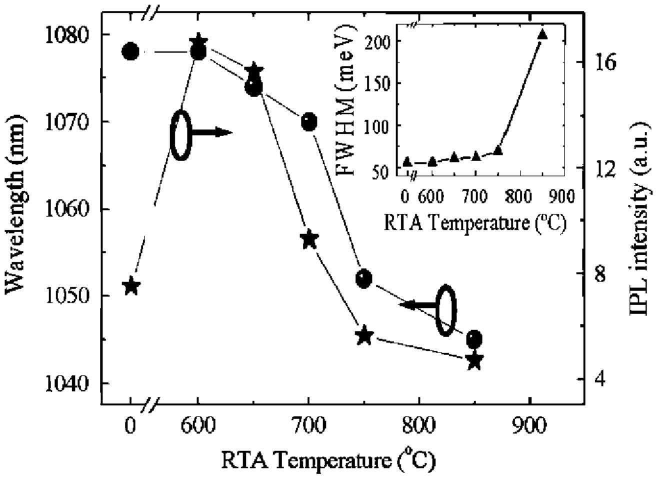

Figure 3. Peak wavelength of PL emission and IPL intensity as a function of the RTA temperatures of annealed InAs/InGaAs DWELL samples. The inset depicts the FWHM of the PL emission as a function of the RTA temperature.

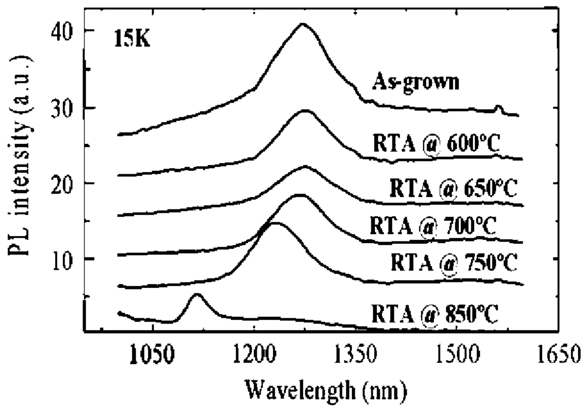

Figure 4. PL spectra at 15 K of as-grown and annealed InAsN/InGaAs DWELL samples.

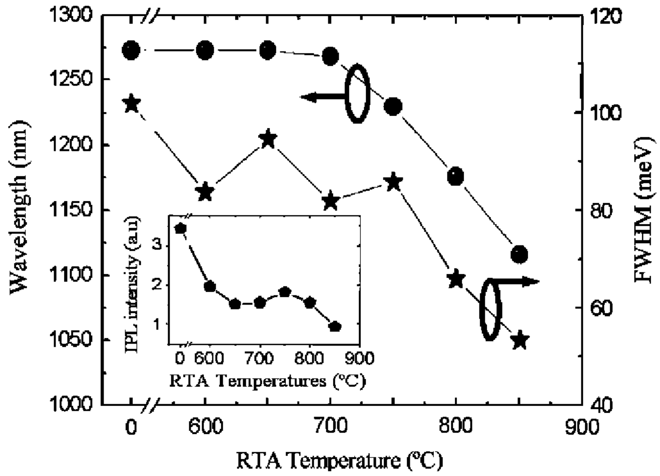

Figure 5. Peak wavelength and FWHM of PL emission as a function of the RTA temperatures of annealed InAsN/InGaAs DWELL samples. In the inset the IPL intensity of the PL emission as function of the RTA temperature is plotted.

the QDs are similar in both types of samples resulting in a comparable structural quality. The mean size and density of these nanostructures are 31 nm and 4.8 QDs cm^{-2} in InAs QDs and 30 nm and 3.7 QDs cm^{-2} in InAsN QDs. However, as can be seen there is greater size dispersion in InAsN QDs (± 8 nm) than in InAs QDs (± 12 nm), which is correlated with the results of the PL measurements.

In figure 3, the emission wavelengths and the IPL intensity of the annealed and as-grown InAs QDs samples are shown as a function of the annealing temperature. It was found that the peak position of PL emission of QDs is hardly blueshifted ($\sim$5 nm) when the annealing temperatures range is between 600 and 700 °C [7]. Nevertheless, a sharp blueshift of the PL peak is observed at 750 °C. Due to the fact that there is no N in the structure, this suggests that In/Ga interdiffusion between the InAs/InGaAs DWELL and the GaAs capping layer is very weak until 700 °C and strong when the RTA temperature reaches 750 °C. At temperatures higher than 750 °C, the blueshift is still present, although in a smoother way. The IPL intensity increases at a RTA temperature of 600 °C, which indicates a clear reduction of the non-radiative centres, and decreases slightly at 650 °C. At RTA temperatures of 700–850 °C, the IPL intensity changes its trend and decreases, implying a degradation of the material quality

As seen in the inset of figure 3, the FWHM has a small change when the sample is annealed between 600 and 800 °C. However, when the sample is annealed at 850 °C, significant broadening of the FWHM can be observed. This suggests that the crystalline quality of the lattice is kept until 800 °C, but at higher temperature the crystalline lattice degrades giving rise to broader PL spectra

Figure 4 shows the PL spectra obtained from the as-grown and the annealed InAsN/InGaAs DWELL samples. The peak wavelength of the PL emission is shifted towards lower energies when the annealing temperature is greater than 700 °C. It was found that while below this temperature the wavelength emission hardly changes (maximum blueshift of 5 nm at 700 °C), the blueshift becomes a little bit more significant at 750 °C (43 nm) and strongly relevant at 850 °C (157 nm). Making a comparison between the PL peak evolution of these samples as a function of the RTA temperature (figure 5), and the behaviour observed previously in the InAs/InGaAs DWELL samples, we found that the effect of post-growth annealing is on both types of structures very close below 750 °C. Nevertheless, if we analyse quantitatively the blueshifts at around 850 °C, we can observe that whereas the PL peak of InAs/InGaAs DWELL is blueshifted only 7 nm from 750 to 850 °C, this blueshift in the case of InAsN/InGaAs DWELL samples is 114 nm. Such a huge difference suggests that additionally to In/Ga interdiffusion, responsible for the blueshift in InAs/InGaAs DWELL samples, other causes lead to the blueshift in the PL spectra of InAsN/InGaAs DWELL samples annealed at 850 °C. These additional causes could be the As–N interdiffusion between the QDs and the barrier and As–N interdiffusion inside the layer of InAsN QDs. These results are in agreement with the observations reported in related to the effect of annealing on the optical properties of InAsN/InGaAs single quantum wells (SQWs). Furthermore, another possible reason could be changes in the local bonding environment of the In–N clusters, as occurring in other dilute nitride systems, such as GaInNAs QWs and QDs

Additionally, figure 5 shows the FWHM as a function of the RTA temperature. The PL FWHM decreases when the annealing temperature increases, but this effect is not significant until 750 °C. As the temperature increases from 750 to 850 °C, the FWHM is reduced sharply. To understand the mechanisms that are responsible for this result, we can observe the previous analysis of the blueshift of PL wavelength and we found that the reduction of FWHM shows the same behavior as the blueshift of the peak wavelength. According to this observation, the effect of annealing on the FWHM could be attributed to a reduction of the non-radiative centres associated with the point defects created by N incorporation. As–N interdiffusion inside the layer of InAsN QDs at 850 °C could help N to accommodate in the crystal lattice thus reducing the point defects (mainly interstitial N). Also, it is possible that N redistribution across the different QDs exists from 600 to 750 °C. However, as the annealing temperature reaches 850 °C, As–N interdiffusion allows the N atoms to accommodate in the crystal lattice and in consequence the effective N concentration in the QDs layer is more homogeneous. This effect also favours the sharp PL FWHM reduction at 850 °C. Therefore, with these results, we can tentatively propose that As–N interdiffusion inside the layer of InAsN QDs is the dominant process responsible for both the sharp blueshift of the peak wavelength of PL emission and the strong reduction of the PL FWHM at 850 °C. In addition to this, bond reorganization of In–N clusters in the QDs may be another possible mechanism for the better optical properties of the annealed samples.

This conclusion is also supported by the fact that the IPL intensity (plotted in the inset of figure 5) of the InAsN/InGaAs DWELL samples improves slightly when the RTA temperature is around 750 °C. This improvement could be also associated with the better quality of the crystal lattice at this temperature due to As–N interdiffusion. The reduction of the IPL intensity from as-grown to 700 °C is still under discussion but it can be related to the possible homogenization of the N content across the different QD structures. It is well known that, using the given growth conditions, the N incorporation is not uniform on the surface of the wafer The as-grown sample probably has QDs containing very low N contents thus providing higher PL intensities. These low RTA temperatures (below 700 °C) help N to redistribute degrading the PL efficiency. Further heating of the sample reduces the point defects resulting in a slight improvement of the IPL intensity.

4. Conclusions

We have investigated the optical effects of post-growth thermal annealing on InAsN/InGaAs DWELL structures by PL measurements, performing a comparison with InAs/InGaAs DWELL structures grown and annealed under the same conditions. A similar blueshift of the peak wavelength and a reduction of the FWHM of the PL emission have been observed in both types of structures until 750 °C. However, at 850 °C these effects are much more pronounced in InAsN QDs due to the additional and dominant roles of the As–N interdiffusion process inside the layer of InAsN QDs and the bond reorganization of In–N clusters in the QDs at this high temperature of annealing.

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